Biocomposites through foam-forming of long fiber suspensions

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ABSTRACT

Replacing synthetic fibers with wood fibers in a thermoplastic polymer matrix is one of the pathways to manufacture carbon-neutral biocomposites. It is known that fibers improve the mechanical properties of composites. However, due to harsh processing conditions in the current technologies, including extrusion and injection molding, the fiber length in the final composite is significantly shorter. Therefore, we coupled foam-forming technology with thermoforming to produce biocomposites with impressive mechanical properties that exceeded the current wood-based thermoplastic composites found in the literature. During foam-forming, the fiber length in the final composite was maintained irrespective of initial fiber consistency and fiber length. Experiments were carried out in both lab and pilot scale. In the laboratory, experiments were mainly carried out to understand the effect of raw material composition on strength properties. Pilot trials were carried out to demonstrate the scalability and to understand the effect of processing conditions to generate a floc-free web with long fibers. The foam-forming consistency ranged from 0.12% to 3 %, which was a significant increase compared to the water-forming process. Initially, foam-formed sheets with varying grammages in the range of 42 g/m^2 to 393 g/m^2 were produced in the pilot machine. The dried foam sheets were then stacked to achieve a grammage of 1200 g/m^2 followed by thermoforming. Foam sheets were made using the following raw materials: a) 1.7 dTex Tencel fiber with the length above 10 mm as long fibers, b) 2 mm wood pulp as short fibers, and c) BiCo fibers comprising polypropylene core and polyethylene sheath or LDPE powder as thermoplastic material. The effect of fiber type, proportion of long fibers, and fiber length on uniformity, strength, and moldability were studied. Visual assessments indicated that the sheet uniformity was good with improved fiber bundle disintegration and reduced flocs even with 20 mm long Tencel fibers. Molding properties were highly dependent on the proportion of fiber, fiber type, amount of thermoplastics, basis weight, density, and the ratio of wood to plastic fibers. In summary, the results indicated that the foam-forming technology enables the manufacturing of long fiber biocomposites with visual and strength properties suitable for packaging, furniture, and automotive applications.

INTRODUCTION

Wood based composites are gaining importance in the past few years, especially in automobile, furniture and household electronics sectors due to the increase in the price of polymers and incentives offered for bio-based products (1). Therefore, researchers are developing thermoplastic composites in which a portion of synthetic fibers are replaced with either short cellulose fibres or long man-made and natural fibers in increasing number of applications (1). Low cost, light weight, origin from renewable resources and wide availability makes the natural and cellulose fibers an attractive alternative to synthetic fibers. In addition to that, from a processing point of view, they possess relatively good mechanical properties such as tensile and flexural modulus, improved surface finish of molded parts composite, less damage to processing equipment, and minimal health hazards (2). Despite the abovementioned advantages, there are a few process-related challenges that exist while handling wood or natural fibers to manufacture composites. In the thermopressing/thermoforming stage of composite manufacturing, the polymer matrix in the composite structure needs to withstand high temperature. The polymer matrix should be selected so that cellulosic fibers do not degrade during processing due to high temperature and the processing temperature should be preferably below 200°C (3). The thermoplastics that soften below this temperature are, for example, polyethylene, polypropylene, polyvinyl chloride, and polystyrene. By selecting one of these hydrophobic polymers as a matrix, compatibility challenges will occur due to hydrophilic cellulosic fibers. Fiber dispersion and fiber-polymer matrix connection related issues are, in many cases, improved by using different coupling agents or using physically or chemically modified fibers (4).

Apart from the limitations with high temperature in processing, handling long fibers in the traditional manufacturing process also poses challenges. Several studies in the literature have shown that an increase in the fiber length improves the strength properties of the composites (5–7). Traditionally, wood plastic composites are made using extrusion, injection molding, compression molding, or thermoforming (pressing) processes. For separate short fibers or sawdust,

extrusion and injection molding are common processes (8,9). In compression molding and thermoforming processes typical for polymer sheets or polymer granules, the addition of natural fibers can be made using needle punched long fiber (e.g., flax) nonwoven-type mats impregnated with polymer and formed into products. In the abovementioned traditional process, due to harsh processing conditions, the long fibers are broken down into shorter fibers limiting the strength properties. One way to retain the fiber length is to utilize the water-forming method, commonly used in paper and nonwoven manufacturing followed by compression molding of material (10). In the water-forming, fibers are dispersed in water in a mixing tank instead of getting chopped down, and therefore, the fiber length is retained. However, the water-forming method consumes large volumes of water, large equipment footprint and the dryer energy consumption to remove the excess moisture from the final product. Recently, foam-forming technology has been used in paper and nonwoven industries to overcome these issues.

Foam-forming technology is seen as one of the potential pathways to manufacture high-performance and resource-efficient cellulose-based materials(11). In foam forming, part of the water from the pulping stage is replaced with an aqueous foam with an air content of 60 to 70%. The fiber-foam is generated by the rapid mixing of water, fiber and foaming agents resulting in uniformly distributed air bubbles with average diameters of less than 100 µm. The fiber-foam is then pumped to forming stage through a headbox, where it is distributed on a wire to form web-like structures. These webs then go through vacuums and dryers to remove the excess moisture from the product. A notable advantage of foam forming is that the air bubbles in foam reduce the fiber flocculation to a greater extent leading to improved formation of the product. Foam forming enables the generation of highly porous web structures by increasing the distance between the fibers. Besides, a wide range of sustainable and value-added products can be manufactured using foam forming based on tailoring the nature of the foaming agent, additives, and fibers dispersed(12)(13)(14)(15). For instance, increasing foam viscosity allows to imbed electronic components to manufacture wearable sensors. On the other hand, replacing the glass fibers with the long cellulose fibers using foam forming(16) results in biocomposites with improved reinforcement properties.

The main objective of this work was to demonstrate the feasibility of manufacturing biocomposites using long fibers through foam-forming technology. Therefore, in this study, lab and pilot scale experiments were carried out using a combination of long fibers, short fibers and thermoplastic component. 1.7 dTex Tencel fiber with length ≥10 mm was used as long fiber. Softwood or hardwood pulp was used as short fiber in lab scale, whereas chemi-thermo mechanical pulp was used as short fiber in the pilot scale. 12 mm long bicomponent fiber or low-density polyethylene powder was used as a thermoplastic component. Lab scale experiments were mainly carried out to understand the effect of fiber type and its proportion on the composite strength. Therefore, the experiments were carried out at a fixed fiber consistency of 3%. In the pilot scale, the focus of the experiments was to understand the influence of processing conditions to generate floc-free structures using long fiber suspensions. Besides, thermoforming trials with three different 3D molds were carried out using the samples from lab and pilot scale to investigate the moldability of long fiber biocomposites.

MATERIALS AND METHODS

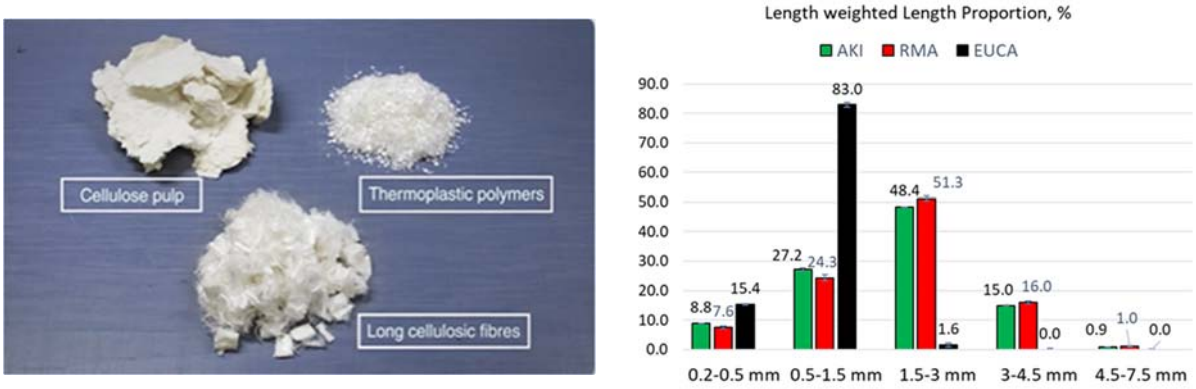
Materials

In this work, 1.7 dTex Tencel fiber (Tencel by Lenzing) with 10 mm length was used as long fibers in the lab scale and 10 mm and 20 mm long fibers were used in the pilot scale. For the lab scale experiments, three types of reinforcing wood fibers were used as short fibres as follows: a) AKI (BSKP – bleached softwood kraft pulp) from Metsä Fibre Äänekoski, b) RMA (BSKP) pulp from Metsä Fibre Rauma, and c) Eucalyptus (hardwood) pulp. In the pilot scale, CTMP pulp was used as short fibers. The properties of the wood fibers were determined with the L&W fiber tester and listed in Table I. Table I shows eucalyptus (hardwood) fiber is shorter and thinner than softwood fibers (AKI and RMA).

Table I: Properties of pulps (AKI, RMA, and EUCA).

Fiber	Mean length (mm)	Mean width (µm)	Fines (%)	Mean shape	Number of large kinks per mm
AKI	1.94	28.6	19.5	80.9	0.37
RMA	2.02	29.2	18.9	82.7	0.24
EUCA	0.74	17.7	21.5	86.7	0.38

Figure 1(a) shows the images of fibers used in the present study. Figure 1(b) shows that the number of fibers longer than 1.5 mm is especially low in the case of eucalyptus pulp. The amount of fines in all pulps was about 20 % (see Table I). The mean shape of RMA fibers was larger than that of AKI fibers, indicating that they were relatively straight compared to AKI fibers. In addition, RMA fibers had a smaller number of large kinks per mm representing a better fiber orientation along the fiber axis.



a b

Figure 1: (a) Snapshot of fibers used in the present study; (b) Fiber length distribution of three short fiber pulps used in lab experiments. The bar color refers to the type of short fiber (green=AKI pulp, red=RMA pulp, and black= eucalyptus pulp)

Two different thermoplastic materials were used in this study. 1) Low-density polyethylene (LDPE) powder from Resinex (LDPE RXP-1005) and 2) 12 mm long polyethylene/polypropylene (PE/PP) bicomponent (BiCo) fiber from FiberVisions (Intraloc 1071). The average particle size of LDPE powder was 350 µm, and the max particle size was 600 µm. The melting point was 108 °C, and the melt flow index at 190 °C was 8 g /10 min. For BiCo fiber, the fiber length was 12 mm, and the linear density (~diameter) was 1.7 dtex. The melting point of the PE and PP were 129-130 °C and 161 °C, respectively. A mixture of sodium dodecyl sulphate (SDS) and Tween 20 (0.3 g/l + 0.3 g/l) from Sigma-Aldrich was used as a foaming agent. SDS is an anionic surfactant with a negatively charged sulphate headgroup and carbon chain (C-12). The molecular weight of SDS is 289 g/mol. Tween 20 (polysorbate-20) is a large non-ionic surfactant of ethoxylated sorbitan molecules with 20 units of polyethylene glycol and a 12-carbon long fatty acid chain. The molecular weight of Tween 20 is 1228 g/mol. Non-ionic surfactant (Tween 20) can function as a dispersion aid and also increases the fiber surface's hydrophobicity.

Composite-Making Procedure: Lab Scale

The production of composites begins by adding all the raw materials to tap water. After adding the foaming agent, the suspension was foamed with a mechanical mixer as shown in Figure 2(a). Foaming took place in a transparent vessel, which enabled the analysis of the foaming phase. The target air content was about 60 % and the fiber consistency was about 3 %. The target grammage was set at 1200 g/m^2, as fiber-reinforced composites with this grammage are commercially available. Foam-formed sheets with a sheet size of 35 cm x 22 cm were made using a hand sheet mold shown in Figure 2(b). When making a hand sheet, the fiber-foam mixture was poured onto the top of the mold, where a funnel directed it to one side. Then, the fiber-foam mixture spread from one end of the mold to the other. This flow was enough to orient fibers in the flow direction. The funnel was then removed, and a plastic cover was placed on top of the foam to create a seal for the vacuum and to enable one-sided dewatering from the bottom. Samples were wet pressed using an L&W wet press (see Figure 2(c)) to remove water and consolidate the wet sheet. Then, samples were dried using a drum dryer shown in Figure 2(d). The samples were thermoformed in the final processing step with a dynamic press device. Dynamic press device (see Figure 2 (e)) has a hydraulic cylinder that clamps the plate attached above to the stationary plate. The maximum pressure in the hydraulic cylinder was 100 bar, and the maximum temperature of pressing plates was 250 °C. The hydraulic cylinder pressure of 95 bar corresponds to ~ 6.2 bar pressure in the 35 cm x 22 cm sample.

Figure 2: Composite-making procedure on a laboratory scale. (a) Foaming of fiber suspension using a mechanical mixer, (b) Sheet formation using foam-forming hand sheet mold, (c) Wet pressing, (d) Drying of samples using a drum dryer. The drying time was 4 hours at 70 °C and (e) Thermoforming using a dynamic press device.

Composite-Making Procedure: Pilot Scale

The pilot scale experiments were carried out at the VTT's SAMPO pilot facility (17), and the schematic illustration of the pilot machine is shown in Figure 3. The pilot machine combines a few unit operations starting with a foaming tank, pumps, inclined headbox, wire section, four vacuum boxes, two dryers, and a sample reeling section. During the experiments, the pilot machine was run in a fourdrinier-former mode and the machine speeds were either 25 or 50 m/min, depending on the basis weight.

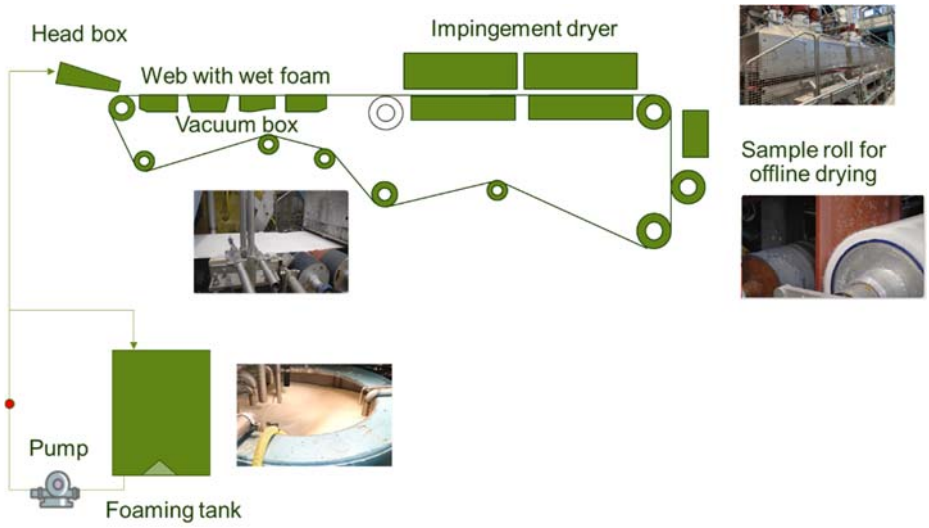


Figure 3: Schematic of pilot-scale setup used to make foam sheet samples for composite making

In the pilot trials with Tencel fiber length of 10 mm, all the raw materials, including the foaming agent, were added into the 5 m^3 foaming tank and then mixed at 1000 RPM between 5 to 15 minutes and recirculated within the foaming tank till the desired foam density was reached. Once the foam was ready, the suspension was pumped to the headbox followed by web generation, water removal and reeling. Due to the machine speed and the limitation on the impingement dryer, the web was not completely dry before reeling the samples. Therefore, the samples for thermopressing were cut from the reel, dried in a drum dryer and thermopressed offline, as shown in Figure 2(d) & (e). For the pilot trials with Tencel fiber length of 20 mm, preliminary trials resulted in rope-like structures leading to

flocs formation as shown in Figure 4. It should be noted that the flocs are not ideal in a final composite structure due to uneven distribution of strength properties. On several occasions, the flocs blocked the pump and headbox. Therefore, for pilot trials with 20 mm long fibers, all the raw materials except for long fibers were added into the foaming tank and then mixed. Once the desired foam density was reached, 20 mm fibers were added and pumped directly to the headbox without recirculation.

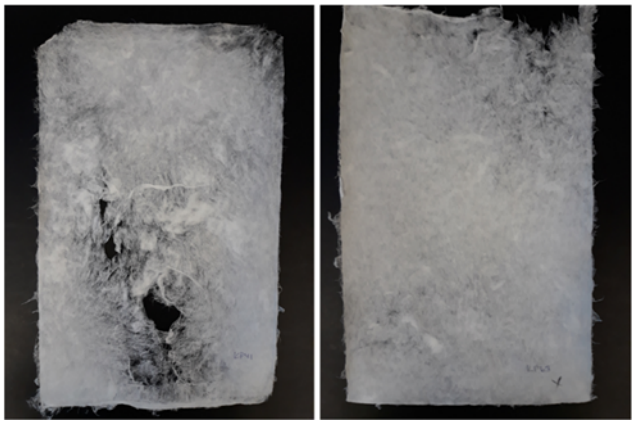


Figure 4: Rope-type flocs for samples with 20 mm long Tencel fibers (with recirculation; 5 to 15 minutes foaming time)

Apart from demonstrating the scalability of composite making procedure through foam-forming, another objective in pilot scale was to make floc-free webs from long fibers. The trial point conditions thereafter included the increase of headbox consistency (starting from 0.2% and ending at 0.5%), reduction of mixing time for the long fibre in pulper (from 60 s to 0s), reduction of mixing energy (rotor speed from 1000 rpm to 500 and 250 rpm) and then increasing the proportion of Tencel fiber (from 20% up to 70%). The foam density varied in a range from 400 to 430 kg/m^3. For the maximum basis weight test, the density was lower ~340 kg/m^3. In total 14 pilot trials were carried out, as listed in Table II.

Table II: Details on the pilot trial points

Trial point	fiber consistency (c), foaming time (t) and impeller speed (rpm)	CTMP %	20 mm Tencel %	12 mm BiCo %
1	Reference : c=0.20%	40	20	40
2	c=0.3%, t=60 sec; 1000 rpm	40	20	40
3	c=0.5%; t = 60 sec; 1000 rpm	40	20	40
4	c=0.3%, t =30 sec; 1000 rpm	40	20	40
5	c= 0.3%, t=30 sec, 500 rpm	40	20	40
6	c= 0.5%, t=30 sec, 500 rpm	40	20	40
7	c= 0.5%, t=0 s, 500 rpm	40	20	40
8	c= 0.3%, t=30 sec, 500 rpm	30	20	50
9	c= 0.3%, t=30 sec, 500 rpm	40	20	40
10	c= 0.3%, t=30 sec, 500 rpm	20	40	40
11	c= 0.3%, t=30 sec, 200 rpm	20	40	40
12	c= 0.3%, t=30 sec, 200 rpm	10	50	40
13	c= 0.3%, t=30 sec, 250 rpm	40	20	40
14	c= 0.3%, t=30 sec, 250 rpm	0	70	30

Composite Strength Measurements

Tensile Strength

Composite tensile strength was determined using the standard for isotropic and orthotropic fiber-reinforced composites (ISO 527-4:2021). The samples were kept in standard conditions (23°C and 50 % relative humidity) for at least five days before testing. The thickness of the samples varied between 2 to 10 mm, and the width was 25 mm. The initial distance between grips during tensile strength measurement was 150 mm, whereas the rate of separation of the gripping jaws was 5 mm/min.

Impact Strength

Impact strength is the capability of the material to withstand a suddenly applied load and is expressed in terms of energy. The toughness of a material can be defined as its ability to absorb energy without fracturing. The impact strength was determined with Charpy impact test where the impact energy required to fracture a sample was measured. In the Charpy test, a three-point bending configuration is used (see Figure 5). The energy transferred to the sample can be determined by comparing the height of the pivoting arm before and after the fracture. Measurements were done according to ISO 179-1:2010(E) standard. The method is suitable for use with fiber-reinforced thermosetting and thermoplastic composites. The preferred dimensions of the specimens are length= 80 mm, width= 10 mm, and thickness= 4 mm. The samples were kept in standard conditions (23°C and 50 % relative humidity) for at least five days before testing.

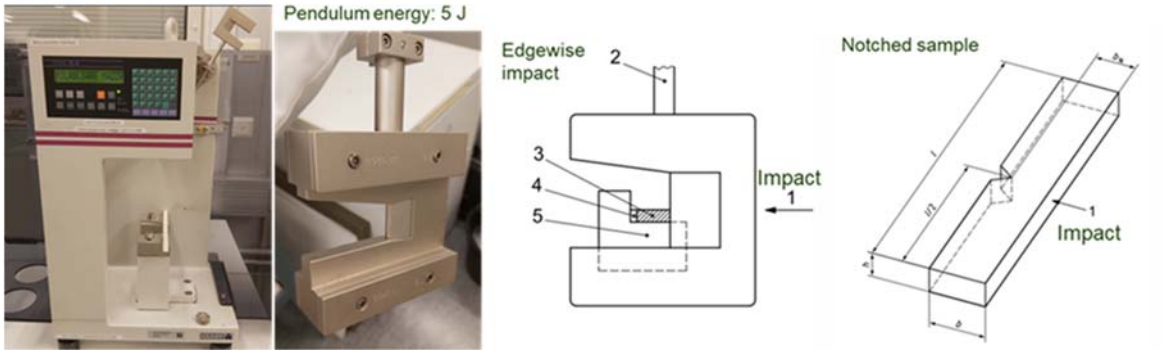


Figure 5: VTT's Charpy impact test device. The impact was directed to the opposite side of the notch. The maximum energy was 5 J.

Thermoforming trials

To study the moldability of the foam-formed composites, thermoforming trials were conducted using MSK Vekomet Hydraulic press as shown in Figure 6(a). Thermoforming trials were performed by pre-heating the sample sheet in the oven for 4-6 minutes, then transferring the sheet to the hydraulic press and pressing the sample. Three different molds were used in the hydraulic press as shown in Figures 6 (b) to (d) to investigate the molding. Foam-formed samples from lab scale were tested only using the cup shape mold and slope type mold due to the limitation on the sample size. Samples from the pilot were tested using all three molds. Thermoforming was carried out mostly on trial-and-error basis to demonstrate the moldability. So, the pressing time, temperature, and pressures used were presented in the results section along with the observations.

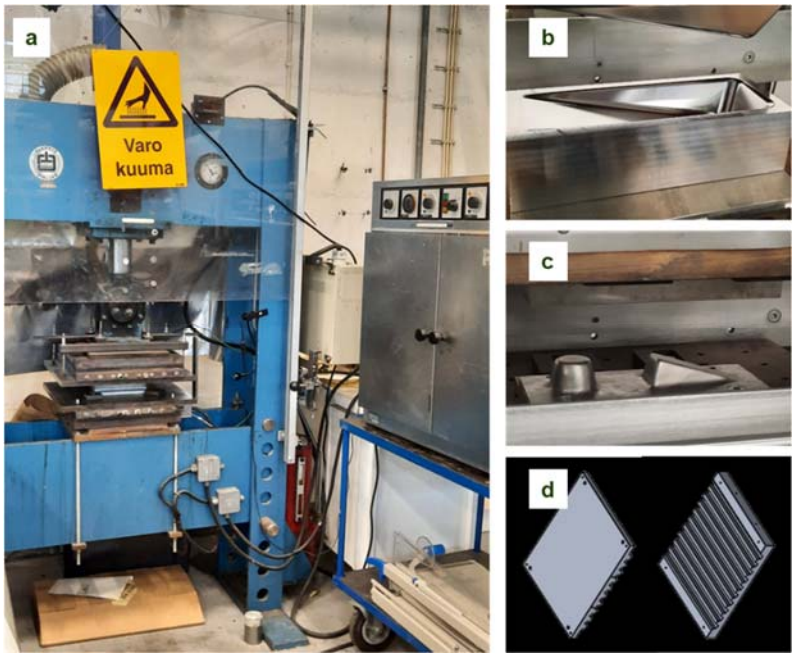


Figure 6: (a) Vekomet hydraulic press and heat oven for thermoforming (b) 20x8 cm slope type mold with 30 mm deepest draw from slope top to bottom (b) cup shape mold and (d) 50x40 cm wave shape mold with 17 mm wave depth

RESULTS AND DISCUSSIONS

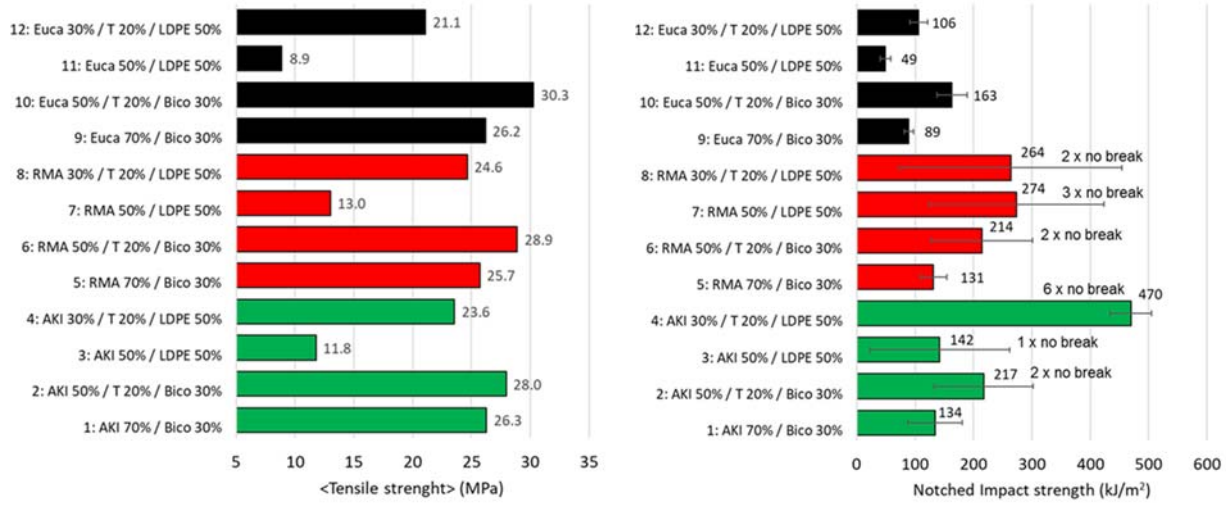
Lab Scale Experiments

Table III: The raw materials of trial points TP1-TP12 and the composite's thickness (µm) and density (kg/m^3) after thermoforming are presented. The table shows the mass percentages of the raw materials.

Trial point	Softwood AKI pulp (%)	Softwood RMA pulp (%)	Hardwood Euca pulp (%)	Regenerated Tencel 10 mm (%)	Thermoplastic BiCo 12 mm (%)	Thermoplastic LDPE (%)	Thickness (µm)	Density (kg/m^3)
1	70	0	0	0	30	0	1870	644
2	50	0	0	20	30	0	1940	622
3	50	0	0	0	0	50	1500	851
4	30	0	0	20	0	50	1300	906
5	0	70	0	0	30	0	1970	611
6	0	50	0	20	30	0	1900	630
7	0	50	0	0	0	50	1420	873
8	0	30	0	20	0	50	1310	918
9	0	0	70	0	30	0	1850	671
10	0	0	50	20	30	0	1920	649
11	0	0	50	0	0	50	1510	830
12	0	0	30	20	0	50	1450	863

The first set of lab experiments was carried out to understand the effect of different fiber proportions and thermoplastic combinations on the strength properties. The list of trial points and measured thickness and density are listed in Table III. When using LDPE powder, the thermoforming temperature setting was 130° C, the pressing time was 2 x 180 s, and the set value for the pressure was 95 bar. The sample was turned 180 degrees between pressings, as the temperature distribution of the plates used for heating was not completely uniform. By turning the sample, we tried to ensure even heating. When using BiCo (PE/PP) fiber, the pressing temperature was 150 °C. The pressure and time were the same as when using LDPE powder. Pressing times were relatively long, as the sample was at room temperature before being placed in the dynamic press device (see Figure 2). The pressing time could be shortened by preheating the sample in an oven. However, in these experiments, no preheating was used. Table III shows that the composites were thinner after thermoforming using LDPE powder. The density of composites made using LDPE powder was about 830-920 kg/m^3. Samples of PE/PP BiCo fiber were thicker and had lower densities. The density of composites made using BiCo fiber was about 610-670 kg/m^3. Adding 10 mm long regenerated fibers slightly decreased the density when using thermoplastic PE/PP BiCo fiber. On the other hand, when using LDPE powder, adding long regenerated fiber reduced the bulk and increased the density. With softwood pulps, the densities were about 40 kg/m^3 higher than when using eucalyptus pulp with LDPE powder.

When the thermoplastic material was used in fiber form i.e, 12 mm BiCO fibers, products with more than 28 MPa tensile strength were manufactured with all three investigated short wood fibers as shown in Figure 7 (a). The share of PE/PP BiCo fiber was only 30 mass-%. In all cases, it was observed that using long regenerated fibers increased the tensile strength. Without long regenerated fibers, composites with a tensile strength of about 26 MPa can be made with 70 mass-% of wood fiber. Using regenerated fiber reduced the strain at break when the BiCo fiber was used as the thermoplastic material. However, adding regenerated fiber increased the strain at break value when using thermoplastic in powder form. The strain at break in softwood samples (AKI or RMA) was higher than in hardwood (eucalyptus) pulp samples. At best, elongations of composites without regenerated fiber were about 11-12 %. Figure 7 (b) shows that the notched impact strengths of several samples were larger than 100 kJ/m^2. In other words, samples were ductile. Composites made of softwood fibers (AKI or RMA) had higher impact strengths than composites made of eucalyptus pulp. The addition of long regenerated fiber increased the impact strength.



(a) (b)

Figure 7: Influence of raw material composition on (a) Tensile strength and (b) Notched impact strength

Generally, the grammage of the samples made in this study has been 1200 g/m^2. This grammage was selected because fiber-reinforced composites with this grammage are commercially available. In the second set of lab experiments, samples with higher grammage (2000 g/m^2) were produced, and we did not face any challenges in making high grammage sheets using foam forming hand sheet mold. Samples with a high grammage allowed samples to be thermoformed to several different thicknesses. In practice, pressing to different thicknesses was carried out by using

metallic spacers of different thicknesses. The samples were thermopressed without spacers or using 5 mm or 7 mm thick spacers. Raw material combinations and the measured thicknesses of samples after thermoforming are listed in Table IV. Table IV shows that the thinnest samples were 1.9 mm, and the thickest samples were more than 7 mm thick after thermoforming. Accordingly, the densities of the samples varied from about 300 to 900 kg/m3 (see Figure 9).

Table IV: The raw materials of trial points and the sample's thickness (µm) after thermoforming are presented. The table shows the mass-% of the raw materials.

Trial point	RMA pulp (%)	Tencel 10 mm (%)	BiCo 12 mm (%)	LDPE (%)	Thickness (µm)
TP6: Ref 1200 gsm	50	20	30	0	1900
TP6: No stopper	50	20	30	0	3520
TP6: 5 mm stopper	50	20	30	0	5420
TP6: 7 m stopper	50	20	30	0	7160
TP8: Ref 1200 gsm	30	20	0	50	1310
TP8: No stopper	30	20	0	50	2400
TP8: 5 mm topper	30	20	0	50	5010
TP8: 7 mm stopper	30	20	0	50	6770

Figure 8 shows that as the density of the samples increases, so does the tensile strength of the samples. When using LDPE powder, the strain at break also increases as the density of the thermoformed sample increases. Figure 9 shows the tensile strength of the samples as a function of the density. The dependence between tensile strength and density differs when using different shapes of thermoplastic particles. In the case of LDPE powder, the individual plastic particles are less than 1 mm in diameter. The fibers, conversely, are thin (12 µm) but long (12 mm) and large in number. Therefore, they can form a more uniform matrix at a low density of composite than powdery particles. Based on Figure 9, it can be assumed that the density of PE/PP BiCo fiber composites should be at least 600 kg/m^3 to reach a tensile strength level of 25 MPa. Achieving a similar strength level when using LDPE powder already requires over 900 kg/m^3 densities.

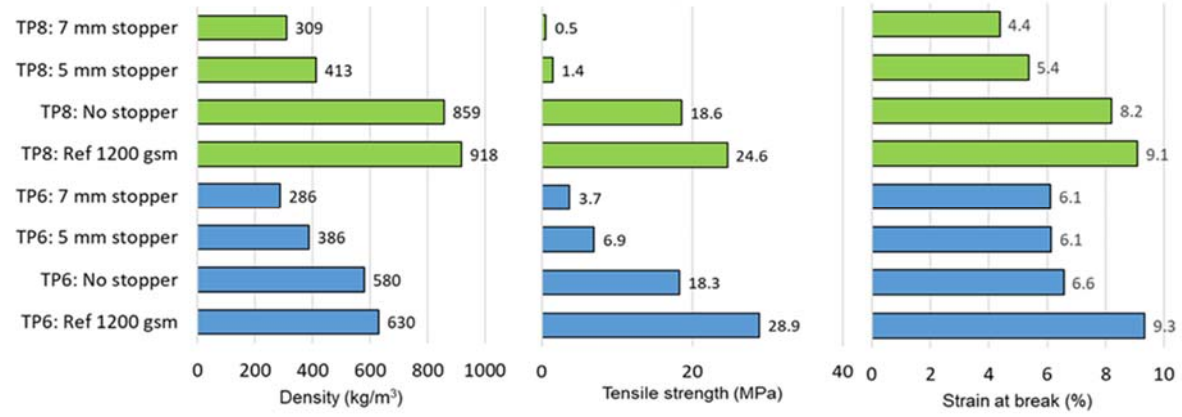


Figure 8: Tensile strength (MPa), strain at break (%). Tensile properties were determined according to ISO 527-4:2021 standard. The bar color refers to the type of thermoplastic material (blue=PE/PP BiCo fiber, green=LDPE powder).

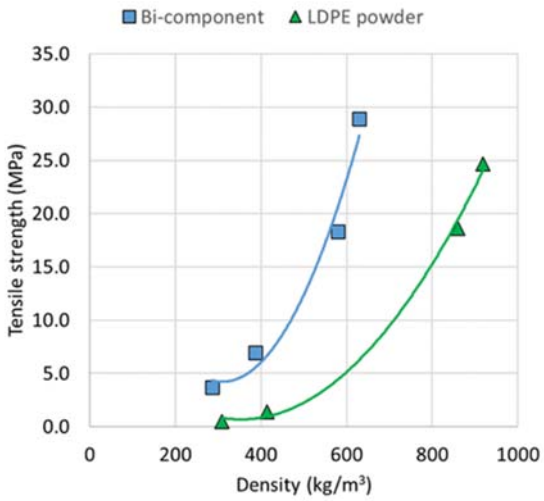


Figure 4: Composite tensile strength (MPa) as a function of density (kg/m^3). Some of the samples were prepared by thermoforming with spacers. Tensile strength was determined according to ISO 527-4:2021 standard. LDPE dosage was 50 mass-%, and PE/PP BiCo dosage was 30 mass-%.

Pilot Trials

The main objective of the pilot trial was to demonstrate the scalability of composite making procedure through foam-forming and to create a uniform web with 20 mm long Tencel fibers without flocs. From the visual inspection, foam-formed sheets from TP5 (see Table II) had the best uniformity where stock consistency was 0.3%, and long fibre mixing was done at an impeller speed of 500 rpm for 30 s. We observed complete dispersion of Tencel fiber bundles and no flocs in the sheets. At high impeller speeds, there were some large rope-type flocs in samples from TP2, TP3 and TP4, indicating flocculation of dispersed fibres in the feed flow. Increasing the headbox consistency or proportion of 20 mm fibre did not clearly impact the sheet uniformity. As the trial focused on the processing of 20 mm fibre, there was no specific target for the basis weight. Basis weight varied between 70 g/m^2 to 190 g/m^2 depending on the variation in consistency. With respect to density, utilisation of non-bonding components resulted in low-density samples around 100 kg/m^3. The lowest values were observed in the samples with the highest Tencel content, and the lowest wood fibre content, respectively. However, the difference between high Tencel and low Tencel content was relatively small, possibly due to the activation of BiCo fibre.

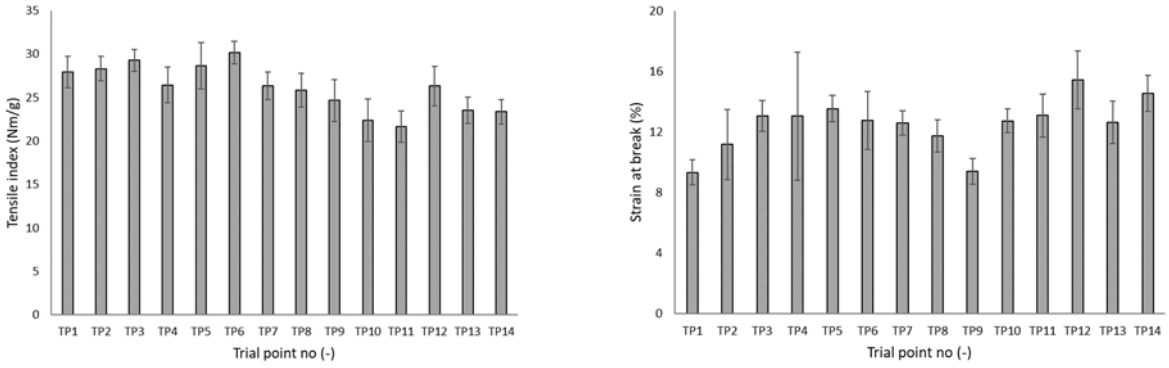


Figure 10: Tensile index and strain at break of the pilot samples. Tensile properties were determined according to ISO 527-4:2021 standard.

The foam-formed sample sheets collected from the reeling section were dried offline in a drum dryer in lab followed by thermopressing. Thermopressed samples were then tested to measure basic structural and in-plane properties shown in Figure 10. Tensile index of the samples varied between 22 and 33 Nm/g, the highest value in the sample where

fiber consistency was 0.5% and the long fibers were mixed for 30 s. Generally, it is considered that the Tencel fibers does not create bonds due to absence of hydroxyl groups leading to lower sheet strength. However, we have not observed any significant reduction in tensile index when the wood fibers were replaced with Tencel fibers. Sheets made from TP14 with 70% of Tencel and 30% of BiCo was at the same in-plane strength level with lower Tencel contents. It can be hypothesised that the presence of BiCo fibers might contributed to the strength properties. Figure 11 shows the Z-strength or thickness directional strength and the influence of Tencel fiber content on the Z-strength. There is a clear dependence on Tencel content i.e, the higher the content, the lower the z-strength. The difference was clearly more significant than in the tensile index. This may be due to the mechanism creating the strength i.e, in tensile mode, the mechanical entanglement of the long fibres increased the strength. In contrast, the inter-fibre bonds contributed more to the strength of thickness directional loading.

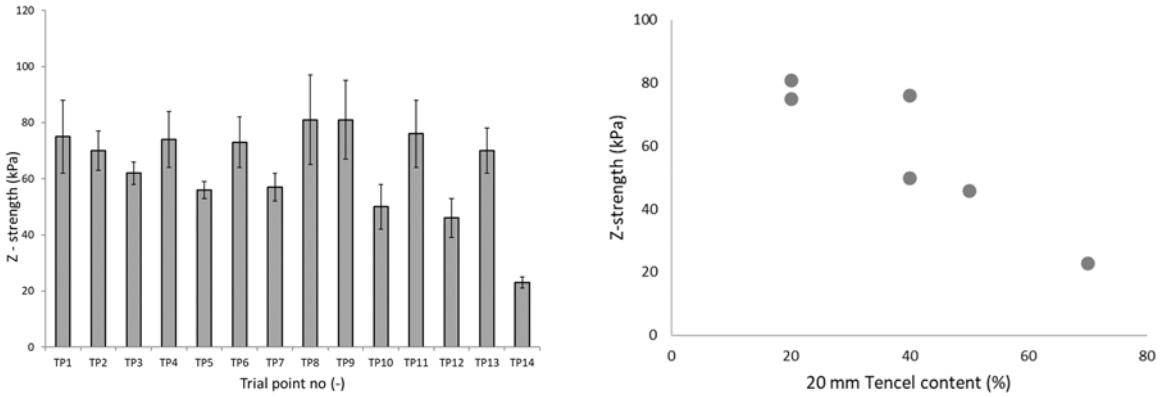


Figure 11: Thickness directional strength and effect of Tencel content on thickness directional strength

Thermoforming Trials

To demonstrate the moldability of the foam-formed biocomposites, thermoforming trials were carried out for samples collected from lab and pilot scale. Figure 12 shows the thermoformed samples made using different mold shapes. It should be noted that to show the ability of the foam forming technique to handle different long fiber types, in Figure 12, we have included samples made from 20 mm long hemp and flax fibers in addition to 20 mm long Tencel fibers.

Figure 12: Thermoformed samples from lab and pilot experiments illustrating the moldability of foam-formed biocomposites (a) cup-shape from lab sample (10 mm long fiber: Tencel), (b) slope shape from lab and pilot sample (20 mm long fiber: Tencel, hemp and flax) and (c) wave shape from pilot sample (20 mm long fiber: Tencel, hemp and flax)

We observed that the samples containing BiCo fibre content of less than 30% resulted in cracks in the cup-shaped mold. In terms of fiber length, moldability improved with the increase in fiber length i.e, 10 mm Tencel fibers resulted in cracks compared to 20 mm long fibers. We observed that the samples with 30% short fiber, 20% long fiber and

50% BiCo fiber formed a soft and plastic-like surface. When the proportion of short fiber was increased above 45%, and BiCo fiber reduced below 35%, samples felt more like cardboard.

Table V: Thermoforming conditions and observations for the stacked samples collected from pilot trials using cup shape mold.

No of sheets	Sample weight (g/m^2)	Raw material composition	Process conditions			Temp. (°C)	Press. (bar)	Observations
			Heating		Pressing			
			Time (min)	Temp. (°C)	Time (min)			
2 sheet	400	CTMP 30%; Tencel 20%; BiCo 50%	6	138	4	53	3 (low)	Low pressure, poor molding
2 sheet	400			146		62	25	Good molding
3 sheet	600			147		52	24	Good molding
4 sheet	800			158		58	29	Good molding
5 sheet	1000			148		54	22	soft on cup and sledge top. Might be due to low pressure
6 sheet	1200			150		55	24	Crack on side of cup and sledge. Feels soft on the top surface. Might be due to low pressure
7 sheet	1400			148		54	30	Cracks on cup and sledge edges. Soft and hairy feel on top surface.
4 sheet	800			152		60	38	Good molding
5 sheet	1000			154		61	133 (high)	Best molding, no creases.
5 sheet	1000	CTMP 46%; Tencel 18%; BiCo 36%		142		61	50	Breaks on cup and sledge top. Might be due to high pressure
5 sheet	1000			154		61	40 Lower pressure	Tears on cup and sledge top, better compared to previous sample
6 sheet	1200			154		61	40	Breaks on cup and sledge top. Might be too thick

For applications such as furniture and automobile interiors, the basis weight of commercial products are in the range of 1500 to 3000 g/m^2. However, due to the inclined headbox setup used in the pilot trials, the basis weight of the samples from the pilot trials was limited between 42 to 393 g/m^2. Therefore, thermopressing of the sheets was used to

combine layers of different basis weights. Depending on the required basis weight, the number of sheets stacked for thermopressing varied from 3 to 20. The stacking approach allowed us to control the (1) basis weight of the sample, (2) manufacture high-weight products by combining low-weight sheets, and (3) multi-layered structures with layers of material with different properties. Details on the thermoforming conditions of the stacked samples and the observations are presented in Table V. We found that alternate stacking i.e, first sheet to the cross-direction and second sheet to the machine direction improved moldability and prevented cracking to a certain extent. Cup shape mold worked for 3 to 4 sheets stacked to basis weight of 600 to 800 g/m^2, whereas 6 sheets with 1200 g/m^2 was too thick to mold using the cup shape mold.

CONCLUSIONS

Several researchers have developed processes to manufacture biocomposites using long fibers. However, in the majority of the cases, due to harsh processing conditions, the benefit of the strength properties due to fiber length was not fully achieved. To address this challenge, we have demonstrated how foam-forming technology enables the manufacturing of biocomposites using long fibers in both lab and pilot scale. A combination of short wood fibers, regenerated cellulose long fibers, and thermoplastics were used to make foam-formed sheets, followed by thermopressing to make composite samples. Tensile and impact strength properties were measured from those samples. In literature, the tensile strength of wood plastic composites has been reported to be between 11-32 MPa, the latter value of which corresponds to a composite made with 50 % plastic. In our lab scale experiments, samples with more than 28 MPa tensile strength were manufactured with all three investigated short wood fibers (bleached softwood kraft pulp and eucalyptus) with 20 mass-% of regenerated fiber. Achieving this level of tensile strength also required that PE/PP BiCo fiber was used as a thermoplastic material. It was also observed that using 10 mm long regenerated fiber increased the tensile and impact strength. The notched impact strengths of several composites were larger than 100 kJ/m^2. Composites made of softwood fibers had higher impact strengths than composites made of eucalyptus pulp. When investigating the effect of the thermoforming pressure on the properties of the composite, it was found that the pressure strongly affects the thickness and density of the composite. In the pilot scale, with 20% of 20 mm long fibre in the stock, the consistency could be increased to 0.5%, representing at least 10-times higher consistency compared to wet-laid processes. In other words, only 10% of the flow volume is needed with foam processing, leading to more energy-efficient process. Also, the disintegration of long fibre in foam was good, and there was no need for separate fibre disintegration with foam as there is in wet-laid process. By visual inspection, the general appearance of the web was uniform and it remained the same even as 20 mm fibre content was increased to 70% of the stock. The produced sheet basis weights ranged from 42 to 393 g/m^2. Thermopressing of 3 to 20 sheets were carried out to generate thick structures with basis weight above 500 g/m^2. In addition, thermoforming trials were carried out to demonstrate the moldability of the foam-formed composites. We found that alternate stacking i.e, the first sheet to the cross-direction and the second sheet to the machine direction improves moldability and prevents cracking to some extent. To verify the ability of foam forming technique to handle different long fiber types, we have made biocomposite samples at pilot scale using 20 mm long hemp and flax fibers in addition to 20 mm long Tencel fibers. The future work will be more focused on fine tuning the raw material composition and processing conditions to generate biocomposites for specific applications including flame retardant and water repellant composites.

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Biocomposites through foam-forming of long fiber suspensions

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Foam-forming technology

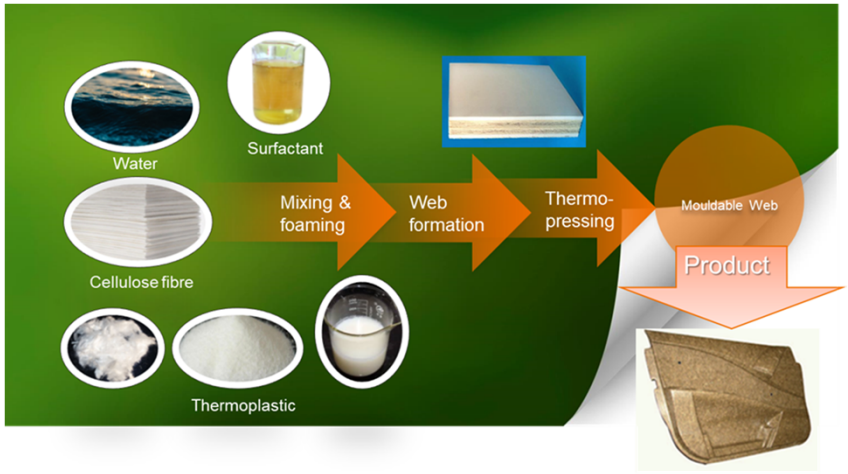
Foam forming technology is a form of paper manufacturing technology mainly used in nonwoven and tissue industry

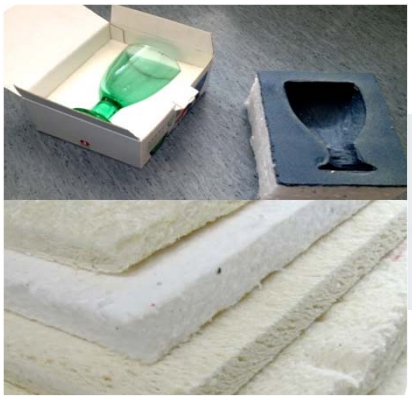
Resource efficient: part of water is replaced by air bubbles in foam-forming

Improved quality & appearance: bubble pockets prevents flocculation, uniform distribution of fibres and additives

Applications: Cushioning, insulation, packaging, hygiene products, composites

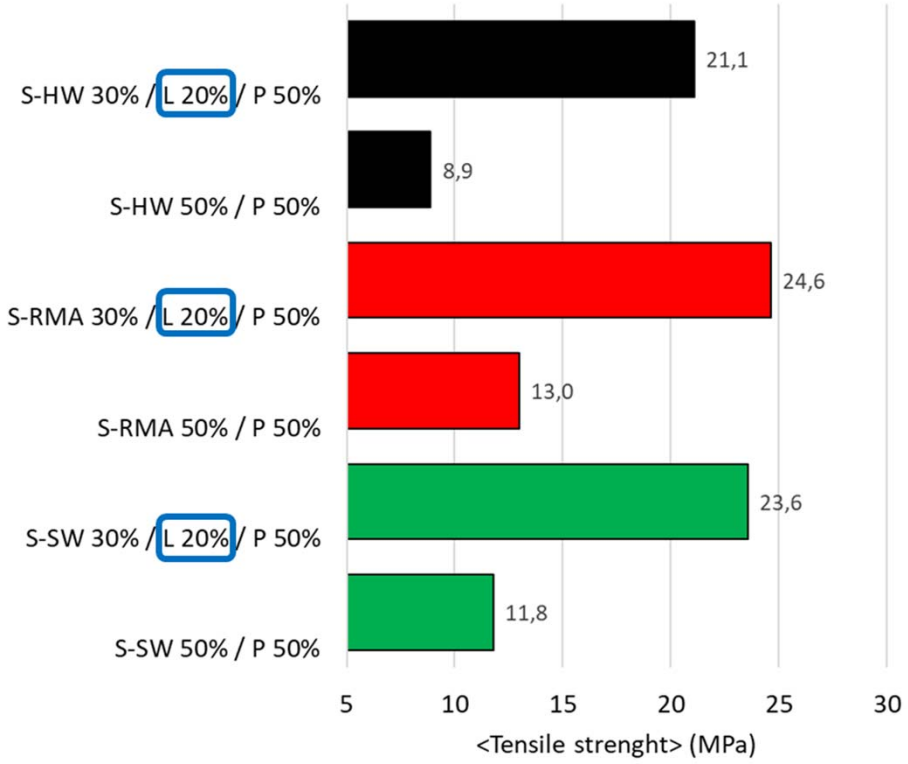
Foam-formed cellulose-based materials **looks like a paper product**, meaning consumers can instantly see that the product if more eco-friendly





Long fibre biocomposites

- **Objective:** To demonstrate the potential of foam-forming technology to manufacture high performance long fibre biocomposites
- Addition of long fibers improves the mechanical performance of the composite
- Compared to extrusion and injection molding methods, in foam-forming fiber length is maintained during the composite manufacturing process
- Wood fibre composition can be increased from 40% to 70% resulting in lower plastic content



P – plastic – LDPE; L – long fiber – Tencel;
S – short fiber – softwood/hardwood

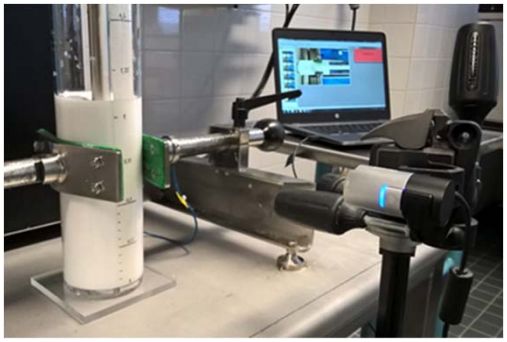
Fibers and additives

Fiber classification	Fiber type	Fiber length
Short fiber	Bleached softwood pulp - Aki	< 3 mm
	Bleached softwood pulp - RMA	
	Eucalyptus pulp	
Long regenerated fiber	Tencel, 1.7 dTex	6 to 10 mm
Long natural fiber	Hemp	5 to 30 mm
	Flax	
Thermoplastic fiber	Bicomponent fiber, PE/PP	12 mm
	Bicomponent fiber, PLA/PLA	
	LDPE powder	350 µm

- A mixture of SDS (sodium dodecyl sulphate, anionic surfactant) and Tween 20 (non-ionic) was used for foaming. Both were usually dosed at 0.3 g/l
- The consistency of fiber suspension was 0.5-5 %. Target grammage 1200 g/m^2 or 2000 g/m^2

Composite making procedure in lab scale

Foaming



- Tap water
- Mechanical mixer
- Air content > 60 %

Sheet formation

Wet pressing

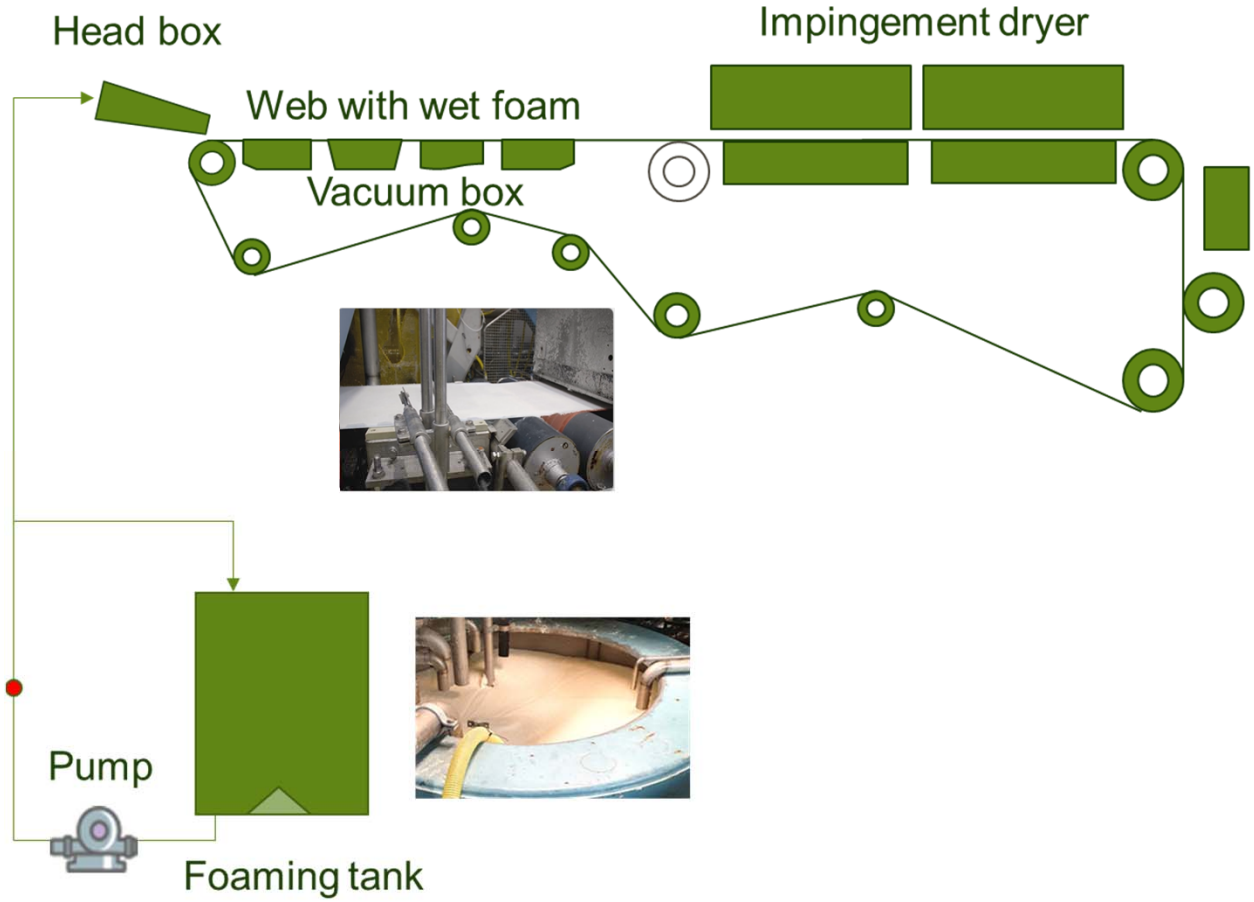
Drying

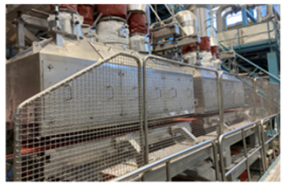
- Drum dryer
- 4 hours at 70 °C.

Thermopressing

- Dynamic press device has a hydraulic cylinder that clamps the plate or the piston attached above to the stationary plate or mold.
- The user can select the compression pressure, pressing time, and plate temperature.
- Sample size: 350 mm x 220 mm
- Pressure: 6.2 bar
- Temperature > Melting point of plastics

Composite making procedure in pilot scale





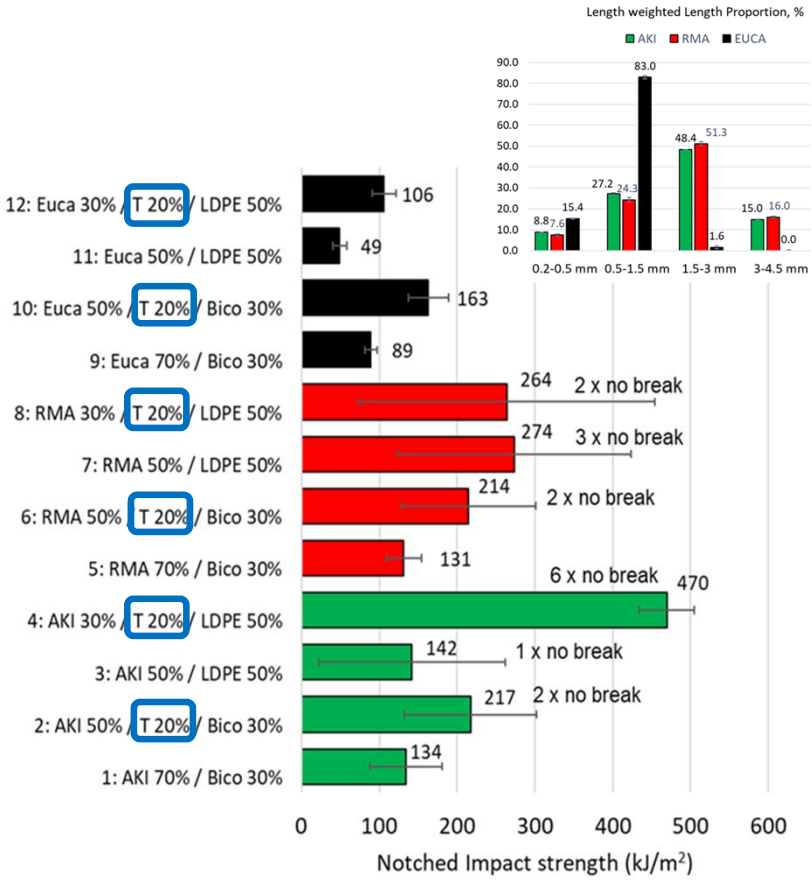
Sample roll for offline drying

Effect of fiber composition

- The notched impact strengths of several samples were >100 kJ/m^2. In some cases, > 200 kJ/m^2

- Composites with more than 28 MPa tensile strength were manufactured with all three investigated short wood fibers with 20 mass-% of regenerated (long) fiber. Achieving this level of tensile strength required that
  - ➢30 mass-% of bi-component fiber was used as a thermoplastic material (PE/PP or PLA/PLA).
  - ➢50 mass-% of LDPE powder was used as a thermoplastic material

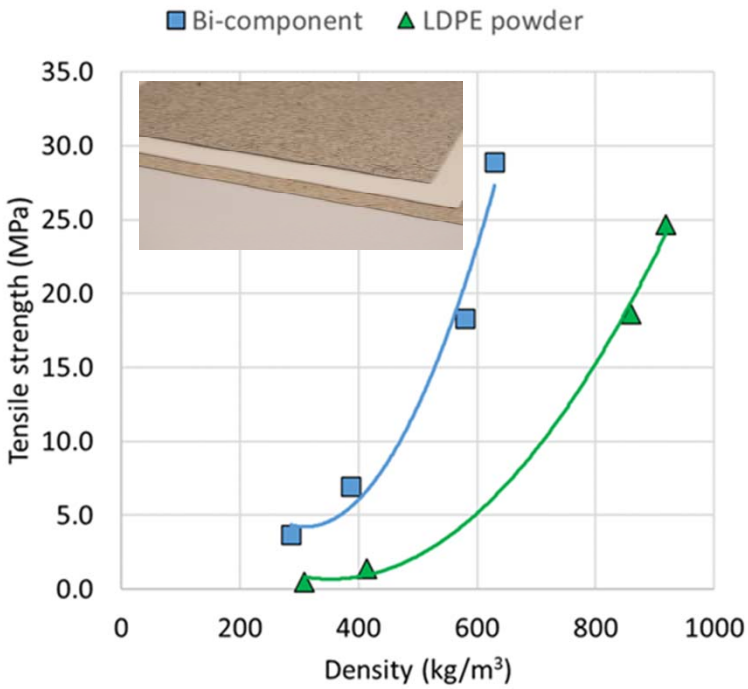
- A tensile strength of up to 35 MPa was achieved using 70 mass-% of lignin-containing CTMP fibers



Euca/AKI/RMA – short fibers; T – Tencel – long fiber; LDPE/Bico – plastic component

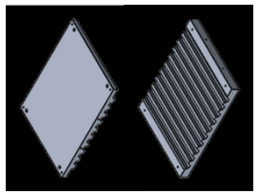
Effect of density & long fiber type

- Sample thickness varied between 1.9 mm, to 7 mm thick after thermoforming and the corresponding densities varied from about 300 to 900 kg/m3
- Individual particles of LDPE powder are less than 1 mm in diameter whereas Bico fibers are thin (12 µm) but long (12 mm) and large in number. Therefore, they can form a more uniform matrix at a low density compared to LDPE powder
- Using a pressure of 24 bar in thermoforming , the tensile strengths of composites were over 60 MPa (densities were about 950 kg/m3)
- Tensile strength of the flax & hemp at were in the range of 24 to 26 Mpa and comparable with Tencel

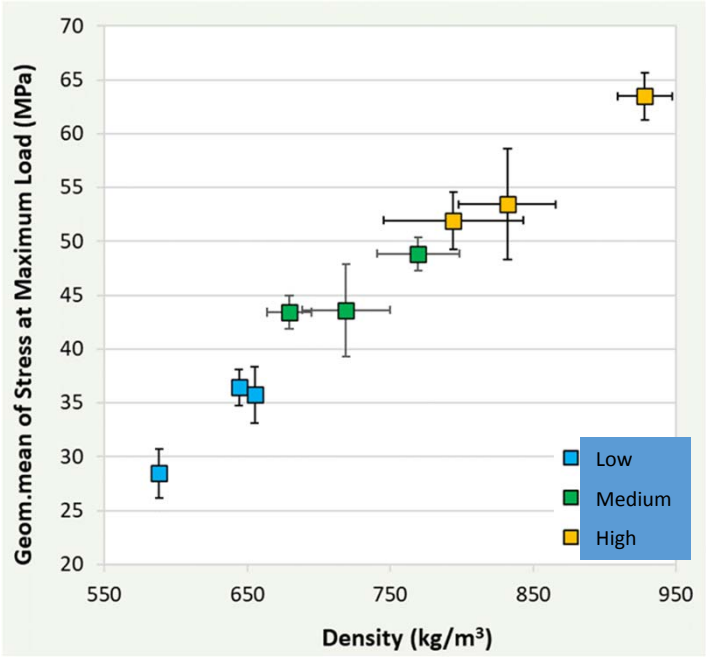


The tensile strength of wood plastic composites has been reported to be between 11-32 MPa, the latter value of which corresponds to a composite made with 50 % plastic

Thermopressing of pilot samples



Parameters: Sample square weight, Long fibre content, Thermoplastic fibre content, Heating time, Temperature, Pressing time, Pressure, Mould temperature and Mould design



Samples with different Tensile strength and product densities were made by changing the process parameters during thermoforming

Summary

- Foam forming combined with thermopressing allows production of high performance biocomposites without loosing the fiber length and reinforcing potential
- 50-70% cellulosic fiber (cellulose, lyocell, viscose, flax, hemp and recycled textiles)
- Foam formed sheets can be stacked to a required thickness of the end product without any adhesives during thermoforming

5000 g/m^2 and 1 cm thick product was successfully demonstrated

- Raw material formulation and thermopressing conditions can be optimized based on the end product

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Thank You

Baranivignesh Prakash, Kirsi Immonen, Timo Lappalainen, Jaakko Asikainen, Miika Nikinmaa, Kristian Salminen
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